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Atmospheric
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Discussions

Interactive comment on "Estimation of volatile organic compound emissions for Europe using data assimilation" *by* M. R. Koohkan et al.

Anonymous Referee #3

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General comments:

This manuscript describes an application of inverse modeling to quantify the extent to which emissions of a selected set of VOCs are accurately represented in existing emission inventories. The authors assimilate "true" observational data from a network of EMEP monitoring sites in an adjoint model based on a 3D air quality model to back-calculate what the emissions should be in order to be consistent with the observations.

This addresses a topic which is highly important to air quality modelers and regulators, but an issue in which little progress has been made to date. We don't really know how good the overall VOC inventories are. It is a difficult problem, as the authors clearly describe in their introduction, and the results of this study show that it is limited by sparse observations, especially for more reactive species – and the ones that may be of most concern for health impacts. The two conclusions I can make from this work are 1.) there may be substantial errors in emissions of some VOCs, which justifies additional work and 2.) if using this technique, a non-truncated Gaussian estimation technique gives the best overall results (?).

The conclusions of this manuscript could be more substantial if it included some discussion of the implications of these results. Does it make sense that propane is underestimated by a factor of 2 or that butene is underestimated by 30%? Why? Is butene underestimated by 30% across-the-board or is it estimated well in some places but poorly in others (and where)? What are the largest potential consequences of these results – why should the reader care?

I cannot fully evaluate the methodology because the manuscript employs terminology (i.e. “hyperparameters”, “gain matrix”, “value screening”) and conventions that are unfamiliar to many readers. It requires more basic explanation of the methodology (maybe putting more of the details in the appendix). Although I try to read sections 2.4 and 2.5, I always end up skipping over these. I hope that other commenters weigh in on the validity of the technique.

The overall presentation could be clarified, with the evaluation of the validity of the assumptions easier to determine with additional explanations. By submitting to ACP, this manuscript strives to appeal to a wider audience (as it should, due to the practical application to a very real modeling problem), therefore the authors should include more text to “state the obvious” – what is the main point being made by Table 9? By Figures 3 and 4? Readers don’t have to fully understand every detail but the figures and tables should communicate something to the reader and they don’t to me. I suspect that these figures and tables could be eliminated.

Specific comments:

There are some variables (and indices) that are not defined, such as T (line 134) or the components of the error covariance matrices I (line 147). Definitions for rs and ms are missing (line 147). The manuscript would benefit from making these definitions very clear up front.

Section 2.1: Errors in the oxidants can mask errors in the emission inventory; the manuscript should present any evidence to support that the CTM is predicting OH, O3 and NO3 that are reasonably accurate.

p. 4, line 97: how are the additional, explicit VOCs "..written in a way that does not affect RACM2?" Are they removed from the lumped species? Do they consume oxidants (as written in Table 1)? A large change in the emissions of any individual VOC could affect the parameters of the lumped species in which that VOC is included, unless you have rederived the lumped species coefficients.

Table 4: Need to define rs and ms in this table (especially since they are not defined well in the text). Include the units of ms and ms+. What is meant by "background"? Does it mean concentrations induced by initial and boundary conditions, or does it mean transported concentrations, or the initial guess in emissions? It is not clear.

p. 8, line 173: I can't find anything in the text giving an indication of the uncertainty in the observations. Since these are used to constrain the emissions, it is important to know how reliable they are.

p. 9, line 206: It seems odd that the boundary conditions of most species are normalized to propane. Need to include some justification that this does not affect the results.

p. 10, line 269, and all simulations using a posteriori emissions; similar to comment on line 97: Did you rederive the emissions of the RACM2 lumped species where applicable or just increase the emissions of the explicit species (I assume the former). Did you need to adjust the parameters for species HC3 in order to account for changes in the

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Discussion Paper

components of HC3 (especially propane and isobutane)?

Figure 7: The discussion of Figure 7 omits the spatial variation in the correction factor, even near to the monitors. While the inversion results (Table 6) imply that emissions for isopentane are close to the a priori emissions, Figure 7 shows a posteriori emissions that vary from 0.10 of the a priori to 5.0 times the a priori. This is important information! To leave the reader with the impression that these emissions are consistent (based on the fact that the overall correction factor is close to 1) is wrong. Why are these four species selected for presentation? Since isobutane and ethene both show substantial error corrections relative to the a priori emissions, these would be more informative than isopentane and o-xylene, and would display the same range of lifetimes.

p. 14, line 342: The comparison with the Cursi et al study raises a lot of questions. Odd that the isoprene inventory is 2.5 times larger than MEGAN, yet your difference in error is only about 20% from the Curci study. I also have not seen a good explanation of error in the assumed yield of HCHO from isoprene, especially in Europe, especially when it is highly spatially and temporally variable, especially based on one chemical mechanism, so I do not agree that the remote sensing option is necessarily more satisfying – it would be preferable to use isoprene directly, as you do.

Table 9: State either in this table of in the text, what the reader should be looking for in the use of ratios in this table. Is a low ratio an indicator of a superior data assimilation system? (see general comment #5)

Technical corrections:

Abstract, line 11: "... the retrieval leads to..." What is the retrieval? Do you mean the a posteriori emissions?

p. 7, line 166:" ...where rs+ and ms+ refer respectively to the standard deviation of the error and of the emission noise..." Which error? Do you really mean the standard deviation of the emission noise?

p. 15, line 410: “For all species the ratio is greater for B1.” Do you mean “For all species the ratio is greater for B1 than B2”?

p. 15, line 414: “The DFS, 4% in the B2 case and 7% is consistent. . .” Which case is 4% and which is 7% or is it 4% and 7% for B2?

References: Line 490: need a better reference for RACM2 – not a conference presentation.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 33219, 2012.

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